Radioactive Krypton Background Evaluation Using Atom Counting

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Abstract

The beta-decay of ^{85}Kr is a significant radioactive background for experiments that use liquified noble gases to search for dark matter and measure the low-energy solar neutrino flux. While there are several proposed methods for reducing Kr levels in these experiments, an independent technique is needed for measuring very low Kr levels. By selectively exciting Kr atoms to a metastable state, capturing them in a magneto-optical trap (MOT), and detecting fluorescence from the trapped atoms, individual Kr atoms can be counted with a high signal-to-noise ratio. This approach could be used to ascertain Kr impurity levels in other noble gases, with an estimated sensitivity of 3×10^{-14}.

1 Introduction

For a variety of ongoing and proposed experiments, the beta-decay of ^{85}Kr is a significant radioactive background. With a current isotopic abundance of

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2.5×10^{-11} in natural Kr[1] and a half-life of 10.76 years, ^{85}Kr gives a beta-decay activity of 3.7×10^5 Bq per kg of natural Kr. In the Earth's atmosphere, with a Kr content of 1.1 ppm, this corresponds to a ^{85}Kr beta decay rate of $1.5\,\mathrm{Bq\,m^{-3}}$. The beta decay of ^{85}Kr extends up to 687 keV, and so Kr impurities are a concern for experiments that are searching for rare events at low energies.

Radioactive background from ^{85}Kr beta decay is a prime concern for experiments that use liquid Xe as a medium for the direct detection of dark matter in the form of Weakly Interacting Massive Particles (WIMPs). In general, experiments designed for direct WIMP searches need to have high sensitivity to nuclear recoils as well as low intrinsic radioactivity. For a more detailed description of WIMPs, their theoretical motivation, and the basic approach to their detection, see Ref. [2]. Liquid Xe is a promising medium for WIMP detection, with a large nucleus, high density, large scintillation yield, and high charge drift velocity.

Several experiments exist or are underway that use liquid Xe as a pure scintillation[3,4] detector or employ a combination of prompt scintillation and proportional scintillation[5,6,7,8,9]. However, since Kr is typically present in research-grade Xe gas at approximately 10 ppm, steps must be taken to remove Kr and its attendant ^{85}Kr decay. At present, Xe can be purchased commercially with a guaranteed Kr level of not more than 20 ppb, but because of the limited sensitivity of conventional measurement of Kr impurities, lower limits cannot be guaranteed. Both the production of Xe with low levels of Kr and the measurement of low levels of Kr in Xe are necessary to evaluate the radioactive background, an essential step for the continued success of Xe as a WIMP detection medium. For example, the XENON detector requires natural Kr

contamination to be 150 ppt or less[10].

Liquid Xe has also been proposed as a detection medium for measuring the $p - p$ solar neutrino flux[11], and in this case the requirement for Kr contamination is even more stringent: $Kr/Xe \leq 4 \times 10^{-15}$. This is similar to the requirement for the CLEAN detector[12,13,14,15], which will use liquid Ne for solar neutrino and WIMP detection. In CLEAN, Kr can be easily removed from Ne by selective adsorption on cold charcoal, but, as in the Xe dark matter detectors, it would be highly useful to have an independent test for Kr contamination. For more information on the scientific motivation for detecting low energy solar neutrinos, see Ref. [16].

Low energy solar experiments using a liquid organic scintillator, such as the Borexino[17,18] and KamLAND[19] experiments (designed to detect solar neutrinos emitted in the $^7Be + e^- \rightarrow {}^8B + \nu_e$ reaction) also require very low Kr levels because ^{85}Kr decay creates events that directly interfere with the signal of interest. However, it is expected that these experiments can be purged of dissolved air (with its attendant Kr content) with pure gaseous nitrogen from liquid nitrogen boiloff. In addition, Kr/N_2 can be determined by removing the N_2 with a getter, then analyzing the remaining noble gas contaminants using a standard mass spectrometer[20].

For Kr contamination in other noble gases such as Ne and Xe, which are promising for low background measurements, it is not practical to separate the Kr chemically. Instead, a possible approach to the measurement of very small Kr quantities in Ne and Xe is the introduction of these gases into a system that can trap and count individual Kr atoms with high efficiency, while simultaneously maintaining a high throughput. The technique of Atomic Trap

Trace Analysis (ATTA), recently developed by Z. T. Lu and collaborators[21], could be used to measure Kr impurities with a sensitivity that is not possible using other currently available methods. This paper evaluates the possibility of using ATTA for measuring trace quantities of Kr in Ne or Xe gases.

The ATTA approach could also be used to measure Ar contamination in Ne or Xe gas, again with the goal of evaluating radioactive background. The isotope of concern in this case is ^{39}Ar, which is produced in the atmosphere by $(n, 2n)$ reactions, has a half-life of 269 years, and has a beta decay endpoint of 565 keV. It is present in the atmosphere at ^{39}Ar/Ar $= (8.1 \pm 0.3) \times 10^{-16}$, as shown by direct low-level counting[22,23] and by accelerator mass spectroscopy[24]. We note that this experimental value conflicts with the theoretical treatment presented by the ICARUS group[25]. Assuming that the experimental values are correct, ^{39}Ar gives a beta-decay activity of 1.0 Bq per kg of natural Ar. In the Earth's atmosphere, with an Ar content of 0.93%, this corresponds to a ^{39}Ar beta decay rate of $1.6 \times 10^{-2} \, \mathrm{Bq \, m^{-3}}$ in air. Though a given quantity of Ar contamination results in much less radioactive background compared to the same quantity of Kr contamination (for example, the CLEAN experiment[15] will require Ar/Ne $\leq 10^{-10}$), it is useful to be able to measure Ar contamination independently. The atom trapping and counting scheme described in this paper could equally be applied to the assay of trace quantities of Ar. Indeed, the transition wavelengths and cooling parameters for Ar and Kr are similar enough that the same apparatus can be used for both.

2 Experimental Approach

Laser cooling and atom trapping methods rely on light pressure forces from photon scattering to reduce the average atomic velocity in a vapor, thus cooling the atoms to microkelvin temperatures[26]. This process offers exceptional isotopic selectivity, as the Doppler shifts due to the motion of the cold atoms are many times smaller than the isotope shifts of the atomic resonance frequencies. Laser cooling has been used to produce ultra-cold samples of roughly 20 different elements, including rare and unstable isotopes ([21,27,28,29]). The ability to selectively manipulate extremely rare isotopes holds great promise for environmental monitoring and geological dating[21,30,31], and the ATTA technique can be extended to measure background impurities in rare-gas samples, which are an important source of systematic error in new neutrino detection experiments. Since the ^{85}Kr/Kr isotopic ratio is well known, a limit on natural Kr derived using such a system would imply a proportionally smaller limit on ^{85}Kr.

The apparatus used for ATTA (Fig. 1) consists of a magneto-optical trap (MOT) loaded from a slowed atomic beam. An atom source produces a beam of atoms moving at thermal velocities (for Kr, $v_{th} \sim 300\,\mathrm{m/s}$ for $T \sim 300\,\mathrm{K}$). These atoms are decelerated by the light force from a counter-propagating laser, and pass through a spatially varying magnetic field that produces a Zeeman shift to compensate the changing Doppler shift as the atoms slow from $300\,\mathrm{m/s}$ to $\sim 1\,\mathrm{m/s}$[32]. Slow atoms from the beam are then captured in the MOT, where a combination of lasers and inhomogeneous magnetic fields trap and further cool the atoms[33]. A typical MOT loaded from an atomic beam will confine 10^7 atoms at a temperature of $\sim 100\,\mu\mathrm{K}$.
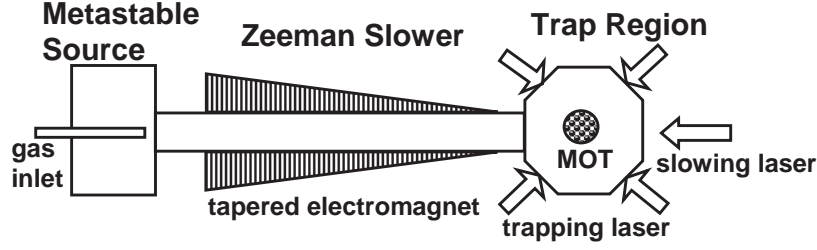
Fig. 1. Schematic of the proposed experimental apparatus for measuring krypton impurities.

The ATTA technique measures isotopic abundances by counting trapped atoms to determine the trap loading rates for an isotope of known abundance and a rare isotope of unknown abundance. The loading rate of the rare isotope is then compared to the loading rate for the more common isotope (measured separately). Assuming the same detection efficiency for the two isotopes, the ratio of loading rates gives the relative abundance. The abundances of ^{81}Kr (1.0×10^{-12}) and ^{85}Kr (1.5×10^{-11}) have been measured by comparing their loading rates to that of ^{83}Kr[21].

Measuring krypton impurities in samples of other rare gases requires measuring the loading rate for ^{84}Kr in a sample of unknown abundance, and comparing to the loading rate from other samples of known abundance. Conventional measurement techniques (mass spectrometry, gas chromatography) can be used to measure Kr impurities at the ppm level. Samples with known Kr abundance can be prepared, and loading rates measured to serve as a reference for comparison with higher purity samples.

The ATTA apparatus is a standard laser cooling system, with two exceptions: In order to trap and cool krypton atoms, we must prepare them in a metastable state, requiring a more complicated atom source than for other atomic species; and in order to detect rare isotopes, we must have a detection system capable

of detecting a single atom in the MOT region.

2.1 Metastable Atom Source

Laser cooling of rare-gas atoms in the ground state is impractical, as the transition wavelengths needed to excite atoms from the ground state to any excited states are prohibitively short ($\leq 126\,\text{nm}$ for Kr) given current laser technology. The lowest excited states of these atoms, however, have extremely long lifetimes (tens of seconds), and serve as an effective ground state for laser cooling. Laser cooling and trapping of atoms in these metastable states has been demonstrated in all of the stable rare-gas species.

In Kr, the metastable state is the $5s[3/2]_2$ state (sometimes called the $^3P_2$ state), with a lifetime of $28\,\text{s}$[38]. Laser cooling is accomplished by exciting the atoms to the $5p[5/2]_3$ state, with a transition wavelength $\lambda = 811.3\,\text{nm}$. Fig. 2 shows some important levels in Kr.

In order to cool and trap Kr, we must produce $5s[3/2]_2$ metastables in the atomic beam source. Ground-state atoms and atoms in other excited states will be unaffected by the cooling and trapping lasers. The excitation can be accomplished either by electron impact, two-photon resonant optical excitation, or two-photon non-resonant optical excitation.

In the electron impact method (Fig. 2a), a plasma discharge is created and maintained using either high-voltage DC current[34] or radio-frequency electromagnetic fields[35]. Gas atoms passing through the source are excited by collisions with electrons in the discharge region. Discharge sources are robust and will work for any atomic species, but they have low efficiency, with
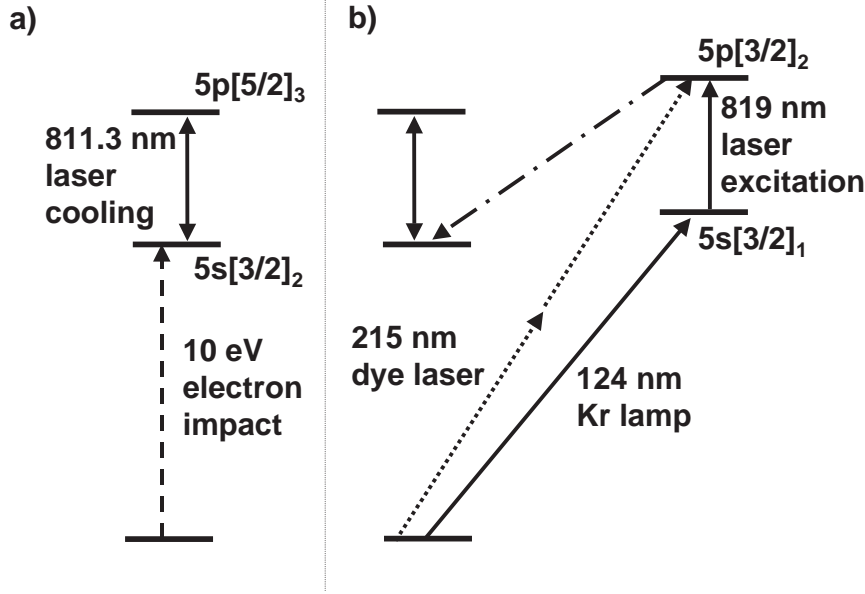
Fig. 2. The atomic levels of Kr. Figure a) shows the energy levels relevant to atomic trapping of Kr*, and figure b) shows the energy levels relevant to resonant and non-resonant optical excitation of Kr*.

metastable atom fractions of $10^{-4} - 10^{-3}$.

The two-photon resonant optical excitation method (Fig 2b) uses a Kr lamp to excite atoms to the $5s[3/2]_1$ state using light at $\lambda = 124$ nm, followed by laser excitation at $\lambda = 819$ nm to the $5p[3/2]_2$ state, which decays to the $5s[3/2]_2$ metastable level with a 77% probability. The remainder of the excited atoms return to the ground state, and can be re-excited.

Because this is a resonant two-photon process, it can, in principle, be more efficient than electron impact. For the present work, it offers the additional advantage of atomic selectivity: only Kr atoms will be excited, while background gas atoms will be left in the ground state. Optical production of Kr* has been demonstrated in a gas cell, and is projected to give a metastable fraction of order 10^{-2} in an atomic beam source[36].

A third possibility is two-photon non-resonant optical excitation, in which two 215 nm photons are absorbed to excite atoms directly to the $5p[3/2]_2$ state, after which they decay with a 77% probability to the $5s[3/2]_2$ metastable level. Because the rate of this excitation scales as the square of the laser intensity, this would best be accomplished with a high-intensity pulsed laser. To allow the interaction with the largest possible number of atoms, the laser can be aimed down the axis of the atomic beam. A pulse repetition rate on the order of 500 Hz should allow each ground-state atom in the beam to interact with the laser. We project a metastable fraction of order 10^{-2} using commercially available pulsed dye laser technology.

The two-photon laser excitation method would simplify the apparatus somewhat, by requiring only a single additional light source. This simplification and the expected increase in excitation efficiency over discharge sources must be weighed against a probable reduction in trap capture efficiency, as atoms excited by the laser near the end of the Zeeman slower may not be decelerated sufficiently to allow their capture in the MOT.

Any of these excitation methods, applied to pure samples of Kr, will produce an atomic beam with a Kr* flux of $10^{13}\,\mathrm{s}^{-1}$ or better. These methods will also work to excite Kr* atoms from impurities in Ne or Xe samples, though discharge sources will also produce Ne* and Xe* metastables, which complicates the collisional dynamics in the sample.

2.2 Single-Atom Detection System

Typical metastable atom sources, using abundant isotopes, give MOT capture rates on the order of $10^7 - 10^8 \, \text{s}^{-1}$, with a lifetime in the MOT of $\sim 1 \, \text{s}$. When working with either rare isotopes or impurities at abundances of 10^{-14}, the capture rate drops to $10^{-3} \, \text{s}^{-1}$ or less, so the average number of trapped atoms will be less than one. In order to determine the abundance of rare isotopes we must have a detection system capable of unambiguously detecting a single trapped atom.

Single-atom detection relies on the fact that atoms trapped in a MOT scatter photons isotropically at a rate of $10^7 \, \text{s}^{-1}$. A fraction of this light ($\sim 1\%$) can be collected using lenses, and detected with an avalanche photodiode (APD). Typical APD photon counting efficiencies are $\sim 25\%$, giving photon count rates of $10^3 - 10^4 \, \text{s}^{-1}$. Contamination by background light is minimized by focussing the collection system on the very small trap volume ($< 1 \, \text{mm}^3$), allowing the fluorescence of a single trapped atom to be easily distinguished. Single-atom detection has been demonstrated for Cs[37] and Kr*[21], with count rates of $10^4 \, \text{s}^{-1}$ above background and S/N~ 40.

2.3 Laser System

In order to place a limit on Kr impurities in other rare gases, it is sufficient to measure the abundance of ^{84}Kr, the most abundant stable isotope (57% in atmospheric samples). ^{84}Kr, like all even atomic mass isotopes of rare gases, has zero nuclear spin, and thus no hyperfine structure. As a result, ^{84}Kr can be cooled and trapped with a single laser frequency, with no repumping lasers

required. The total laser power required for trapping and cooling is of order $100\,\mathrm{mW}$, and can be obtained using grating-stabilized diode lasers[38].

2.4 Collisions and Background

Inter-species collisions are an important potential complication for measurements of Kr impurities in Ne or Xe samples. There are two reactions of particular importance:

$$\mathrm{Ne^* + Kr \rightarrow Ne + Kr^+} \tag{1}$$
$$\mathrm{Kr^* + Xe \rightarrow Kr + Xe^*} \tag{2}$$

where $\mathrm{Ne^*}$ represents the $3s[3/2]_2$ metastable state, and $\mathrm{Xe^*}$ refers to any one of several possible excited states in Xe. The internal energy of an atom in a metastable state can be transferred to a ground-state atom during a collision. These energy transfer reactions can change the excitation efficiency in the source, and thus the overall loading rate for $\mathrm{Kr^*}$.

Eq. 1 describes Penning ionization of ground-state krypton in collisions with neon metastables, which have an internal energy ($16.6\,\mathrm{eV}$) greater than the ionization potential for krypton ($14.0\,\mathrm{eV}$). Room temperature cross sections for this reaction are of order $10^{-15}\,\mathrm{cm^2}$[39], giving a mean free path for $\mathrm{Kr^*}$ in Ne of several meters for the discharge parameters of Ref. [35]. Moreover, this reaction can be entirely avoided by using an optical excitation source, where only Kr atoms are excited. The internal energy of $\mathrm{Kr^*}$ ($9.9\,\mathrm{eV}$) is too low to excite any state in ground-state neon, so the effect of collisions between excited Kr and ground-state Ne should be negligible.

The $\mathrm{Kr^*}$-Xe collisions described by Eq. 2 are more problematic. In this case,

the internal energy of the Kr* metastable is transferred to a ground-state Xe atom, populating high-lying excited states in Xe, which decay rapidly. As energy is transferred from Kr to Xe, this reaction cannot be completely avoided by optical excitation.

Room temperature cross sections for these Kr*-Xe collisions are of order $10^{-14}\,\mathrm{cm}^2$[40,41], giving a mean free path for Kr* in Xe under the conditions of Ref. [35] of $\sim 1\,\mathrm{cm}$. This will produce a significant reduction in the metastable production efficiency. This problem may be alleviated somewhat by designing the source so that metastables are created in a region of very low gas density, but collisional quenching in the source will be a major factor limiting the sensitivity of ATTA measurements in Xe.

2.5   Sample Contamination

Outgassing of Kr atoms from the walls of the vacuum chamber could in principle limit the accuracy of ATTA for this application. The fact that the atoms must be in the metastable state in order to be trapped and detected means that only outgassing in the source region, where the atoms have a chance of being excited to the metastable state, needs to be considered. Outgassing in the MOT chamber will consist of ground-state atoms, which will not be trapped, and thus will not contribute to the abundance measurement.

While relatively little is known about noble gas outgassing from metals at room temperature, both solubility and diffusion for noble gases are near zero in metals. Hence outgassing is dominated by nitrogen, carbon monoxide, hydrogen, water, and hydrocarbons. Some experiments measuring Kr isotope

ratios in meteorites[42] require low Kr outgassing in noble gas mass spectrometers, which are mostly metal with some attached glassware and/or ceramic pieces. The standard final cleanup of these systems involves bake out under vacuum of the complete assembly at a temperature of 350 C. The background is dominated by the contributions from the heated parts during the preparation of the samples, the operation of the valves, and the non-metallic parts of the system[43]. A ^{82}Kr outgassing rate of 5×10^{-15} cc STP is typical for outgassing times of the order of 60 minutes and chamber areas of the order of 1000 cm.

Based on this outgassing rate, we can expect 2×10^{1} ^{84}Kr atoms/s outgassed into a source chamber of area $100 \, \text{cm}^2$[35]. Given a sample injection rate of 7×10^{16} atoms/s, this gives a Kr background of 3×10^{-16} Kr/Ne or Kr/Xe, assuming that Kr atoms from the walls are excited in the source and captured in the trap with equal efficiency as Kr atoms from the sample. In a future ATTA setup for measuring Kr/Ne or Kr/Xe, further improvements in Kr outgassing are likely, assuming sustained baking of the vacuum chamber and careful choice of materials.

An important factor that may complicate this analysis is the embedding of Kr$^+$ ions in the walls during the operation of a discharge source. These atoms can emerge from the walls later, either through normal outgassing, or by being driven out by later ion impacts. Du *et al.*[44] report this as a source of cross-sample contamination in experiments to measure isotopic abundances in atmospheric samples, accounting for more than 1% of their Kr signal. This "memory effect" can be minimized by aligning and testing the trap system with pure argon (the laser cooling wavelengths for Ar* and Kr* differ by less than 1 nm, and thus use the same lasers and optics). Even so, it is potentially a

serious problem for a system using a discharge source when switching between reference samples with relatively high Kr abundance, and the ultra-pure samples of interest. The effect can also be avoided by using an optical excitation source, which will not produce ions or embed atoms in the source walls.

3   Discussion

The ATTA method should allow highly sensitive detection of Kr atoms in Ne or Xe gases. Lu *et al.* claim a capture efficiency in their trap of about 1 part in 10^7, fairly typical for beam loading systems[21]. Their basic atom source consumes 7×10^{16} atoms per second, which would give a trap loading rate of about 4×10^9 of the most abundant isotope (^{84}Kr, 57%). Making the simple approximation that the Kr capture efficiency would be the same in a different gas, one gets a sensitivity of 3×10^{-14} in 3 hours of integration. This assumes a discharge source with 10^{-4} metastable fraction, as was used in [21]. If optical excitation is used, one could achieve a significantly higher metastable fraction, potentially enhancing the Kr capture efficiency and loading rate.

Based on the above simple calculations, we estimate that trace quantities of Kr in Ne and Xe gases could be detected using Atom Trap Trace Analysis. Sensitivities as high as 1 part in 3×10^{14} should be possible using a simple discharge source, and even higher sensitivities should be possible using optical excitation of the Kr. Similar sensitivities should be possible for measuring trace impurities of Ar. This technique may prove valuable for evaluating radioactive contamination in experiments using condensed noble gases for the detection of low energy neutrinos or dark matter.

4 Acknowledgements

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